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FISSION PRODUCT HOLDUP IN GRAPHITE

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INTRODUCTION

Multicomponent time-dependent concentration diffusion and radioactive decay of isotopic species is an important aspect of fission product migration and release from fuel particles and fuel elements in a High Temperature Gas-Cooled Reactor (HTGR). After fission products escape from a fuel particle in an HTGR, it is still necessary for them to diffuse across the graphite web of a fuel block to a coolant hole before they can be entrained in the primary coolant. The time required for a given fission product species to diffuse across the graphite web has a direct influence on the time-dependent release associated with a significant increase in the power/flow ratio. The main purpose of this paper is to present the results of a study of the holdup time in graphite of Sr as a function of the diffusion constants. The study employs a newly-developed multicomponent time-dependent diffusion and decay code called DASH.¹

Analysis methods for solving the type of problem discussed here are well known,² and some applications to fission product decay and diffusion in HTGRs have appeared in the literature.³ However, the methods employed are often subject to time step limitations, and the effects of decay are not adequately handled. The DASH code uses a one dimensional spatial discretization for the diffusion operator and an analytic matrix operator method to remove the time dependence. The matrix operator method is described in Ref. 4 where the method was applied to the modeling of the consequences of a fission product release into the primary coolant of an HTGR. Comparisons of the solutions given by DASH with a number of analytic solutions have been made, and in all instances considered the agreement with analytical solutions is excellent and limited only by the inaccuracy inherent in the spatial discretization.¹

ANALYTICAL MODEL

The differential equation governing time-dependent multicomponent diffusion with radioactive decay is given by

$$\frac{\partial \vec{C}}{\partial t} = \nabla \cdot D \nabla \vec{C} - \lambda \vec{C} + \vec{S} \quad (1)$$

where \vec{C} is an n-component column vector representing isotopic concentrations (atoms/cm³), D is an nxn positive definite diffusion matrix (cm²/s), λ is the decay matrix (s⁻¹), and \vec{S} is an

n-component source vector (atoms/cm³-s). In DASH Eq. (1) is solved in slab, cylindrical, or spherical geometries subject to specified initial and boundary conditions on \vec{C} .

The spatial derivatives in Eq. (1) can be removed by standard finite difference or finite element numerical techniques using a spatial mesh. In Ref. (1) a finite difference approximation for the diffusion operator is obtained for a nonuniform mesh in slab, cylindrical, or spherical geometries. For m cells the spatially discretized equations that result are of the form

$$[v] \frac{d\{\vec{C}\}}{dt} = [A] \{\vec{C}\} + [v] \{\vec{S}\} \quad (2)$$

where $\{\vec{C}\}$ and $\{\vec{S}\}$ represent mxn vectors

$$\{\vec{C}\} = \begin{bmatrix} \vec{C}_1 \\ \vec{C}_2 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ \vec{C}_m \end{bmatrix}, \quad \{\vec{S}\} = \begin{bmatrix} \vec{S}_1 \\ \vec{S}_2 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ \vec{S}_m \end{bmatrix} \quad (3)$$

and $[v]$ and $[A]$ are volume and diffusion-decay matrices of the form,

$$[v] = \begin{bmatrix} v_1 & & & & \\ & \cdot & & & \\ & & \cdot & & \\ & & & 0 & \\ 0 & & & & \cdot \\ & & & & & v_m \end{bmatrix}, \quad [A] = \begin{bmatrix} K_1 & A_1 & & & \\ A_1 & K_2 & A_2 & & \\ & \cdot & \cdot & \cdot & \\ & & \cdot & \cdot & A_{m-1} \\ & & & A_{m-1} & K_m \end{bmatrix} \quad (4)$$

where V_i , $i = 1, m$ are $n \times n$ diagonal matrices that depend on the cell volumes; K_i , $i = 1, m$ are $n \times n$ matrices that depend on both the diffusivity and decay matrices; and A_i , $i = 1, m-1$ are $n \times n$ matrices depending on the diffusivity matrix.

Although Eq. (2) can be solved by standard time differencing techniques, such techniques were not used because of constraints on time step size to ensure numerical stability and because of inaccuracies that would arise in calculating the decay of the isotopic chains when large differences in decay constants are present. Instead, an operator method was used. By standard techniques^{1,4} the solution of Eq. (2) is

$$\{\vec{C}(t)\} = [V]^{-1} e^{[B]t} [V] \{\vec{C}(0)\} + [V]^{-1} t D([B]t) [V] \{\vec{S}\} \quad (5)$$

where

$$[B] = [A] [V]^{-1}$$

and

$$D([B]t) = \sum_{p=0}^{\infty} \frac{([B]t)^p}{(p+1)!} \quad (6)$$

ACCURACY EVALUATION

Although no experimental validation of DASH has been conducted, a substantial number of comparisons have been made to published analytic solutions. No attempt has been made to make all the possible comparisons but a sufficient number of problems have been compared to establish confidence in the DASH methodology. For the problems considered, the observed errors are of the magnitude one would expect from a spatial finite differencing technique. These comparisons are discussed in detail in Ref. 1. The test problem discussed below was chosen because it points up certain unique features of the code.

The simplest problem type to utilize the full capabilities of the DASH code is the one material, one specie problem with concentration diffusion and radioactive decay. Although the one-dimensional geometry in the code permits the evaluation of problems involving an infinite slab, an infinite solid or hollow cylinder, and a solid or hollow sphere we have taken the slab geometry as an example. The analytic solution for comparison is taken from Crank.⁵ Since the published results are for concentration diffusion without radioactive decay, a transformation developed by Danckwerts⁶ was used to extend these results for time-dependent concentration diffusion to also handle radioactive decay. Danckwerts transformation states that

$$C = \lambda \int_0^t C' e^{-\lambda t'} dt' + C' e^{-\lambda t} \quad (7)$$

where λ is the radioactive decay constant (1/s), C' is the diffusion solution without radioactive decay (atoms/cm³), t is the evaluation time

(s), and C is the solution with both diffusion and decay (atoms/cm³). This transformation is valid for an initial concentration of zero, and boundary conditions of either surface-saturation or surface resistance.

The analytic solution for time-dependent concentration diffusion in a slab ($0 \leq x \leq \ell$) with a uniform initial distribution and different saturated surface concentrations is⁵

$$C = C_1 + (C_2 - C_1) \frac{x}{\ell} + \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{C_2 \cos(n\pi) - C_1}{n} \sin \frac{n\pi x}{\ell} \exp \left(\frac{-Dn^2\pi^2 t}{\ell^2} \right) + \frac{4C_0}{\pi} \sum_{m=0}^{\infty} \frac{1}{2m+1} \sin \left(\frac{(2m+1)\pi x}{\ell} \right) \exp \left(\frac{-D(2m+1)^2\pi^2 t}{\ell^2} \right) \quad (8)$$

where C_0 is the initial uniform concentration, C_1 is the surface concentration at $x = 0$, C_2 is the surface concentration at $x = \ell$, D is the diffusion coefficient, and t is the evaluation time.

A simple one material, one specie infinite slab problem has been defined which can be solved by both Eqs. (7) and (8) and by DASH. The test problem was solved analytically at 27 space points at 5 different times. The DASH solution was for the same 5 times using 25 mesh cells. The maximum error observed occurred during the first time step, 0.1 days, at the center of the slab and had a magnitude of 0.28%. The results are compared in Fig. 1. The figure resolution is such that the analytic and analytic-numerical, DASH, results fall on top of each other.

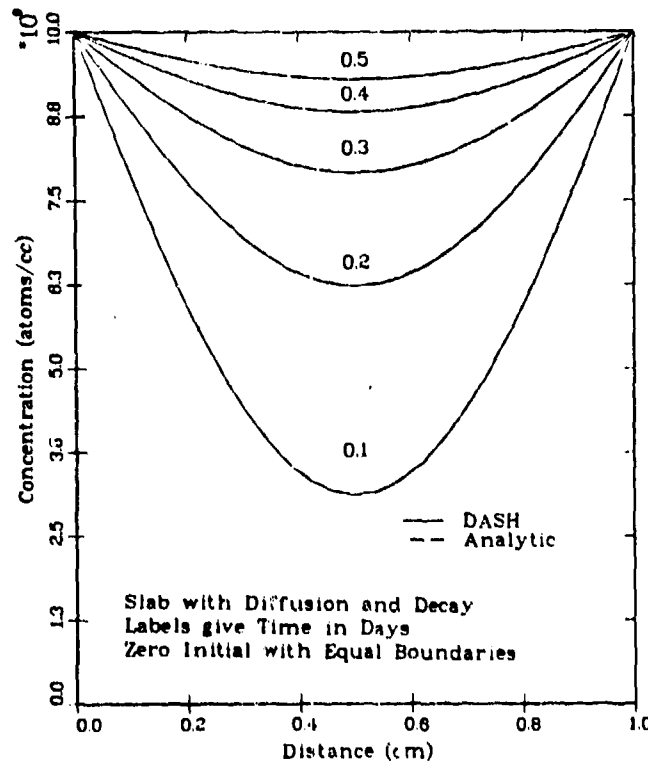
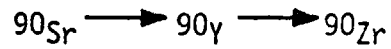


Fig. 1. Slab validation problem results.

HOLDUP OF ^{90}Sr BY GRAPHITE

A parameter study of the release and diffusion-decay of isotopes of strontium in a simplified one-dimensional slab model of an HTGR core block has been carried out. A typical element of the core block and the coolant hole was modeled as shown in Fig. 2; the dimensions of each region were taken from Ref. (3).

The decay chain used for the test problem is



with yields and decay constants shown in Table I.

TABLE I

Isotope	Yield %	Decay Constant/s
^{90}Sr	5.77	$7.844 \text{ E-}10$
^{90}Y	5.77	$2.994 \text{ E-}6$
^{90}Zr	0.0	$1.0 \text{ E-}20$

The boundary conditions used are reflection at $x = 0$, zero concentration at $x = 1.05$.

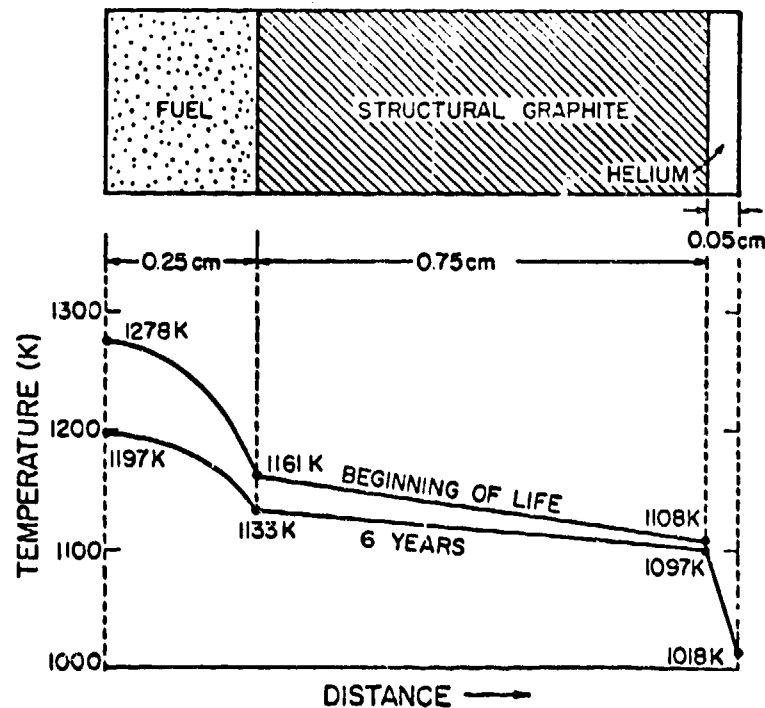


Fig. 2. Fuel-graphite-helium calculational model and beginning of life and six year temperature profiles.

The approach is to use data from the work of Appel and Roos³ and calculate the distribution of the isotopes of this decay chain in the fuel matrix and structural graphite. The source term for ⁹⁰Sr is taken to be 7.3×10^9 atoms/cm³ as given in Ref. 3. The source terms for the other isotopes in the chain are taken in proportion to the yields of Table I.

The temperatures changes from the beginning to the end of the calculation (six years duration) are shown in Fig. 2. Temperatures at intermediate times are calculated by linear interpolation.

Data is given in Ref. 7 for the diagonal terms of the 3x3 diffusion matrix for the three species making up this problem. For the Arrhenius representation

$$-\log_{10} D = A + 1000 B/T$$

the coefficients A and B are given in Table II. The data was taken from Ref. 7.

TABLE II
DIFFUSION COEFFICIENT PARAMETERS

SPECIE	A	B
⁹⁰ Sr	0.34	6.5
⁹⁰ Y	0.74	14.2
⁹⁰ Zr	1.19	22.8

Appel and Roos³ assume that the concentration of ⁹⁰Sr drops by a factor of 300 at the fuel graphite interface corresponding to the distribution coefficient between the two substances. This is handled in DASH by putting a small region (10^{-8} cm thick) at the boundary and adjusting the diffusion coefficient of the region introduced until the ratio of ⁹⁰Sr concentrations is 300. Except for this boundary region, the mesh spacing is taken as 0.05 cm throughout.

The result of the calculation was that a nonmonotonic behavior of concentration with time was observed. This was caused by the competition between an increase caused by the presence of the source over the entire calculational time and a decrease caused by the lower temperatures at later times. When the temperatures were held constant over the 6 year calculational period, the results were not very illuminating as a steady state concentration profile was reached in about one year.

A more realistic treatment of the source⁸ allows for an increased source strength in later years caused by an increase in fuel particle failure rates. We assumed that the initial source ($S_0 = 7.3 \times 10^9$ atoms/cm³) increases with time such that S_0 is used for the first year, $2S_0$ for the second year, $3S_0$ for the third year, etc. Numerical results for ⁹⁰Sr concentration are shown in Fig. 3 and the ⁹⁰Y concentration profiles are shown in Fig. 4. Comparison of the

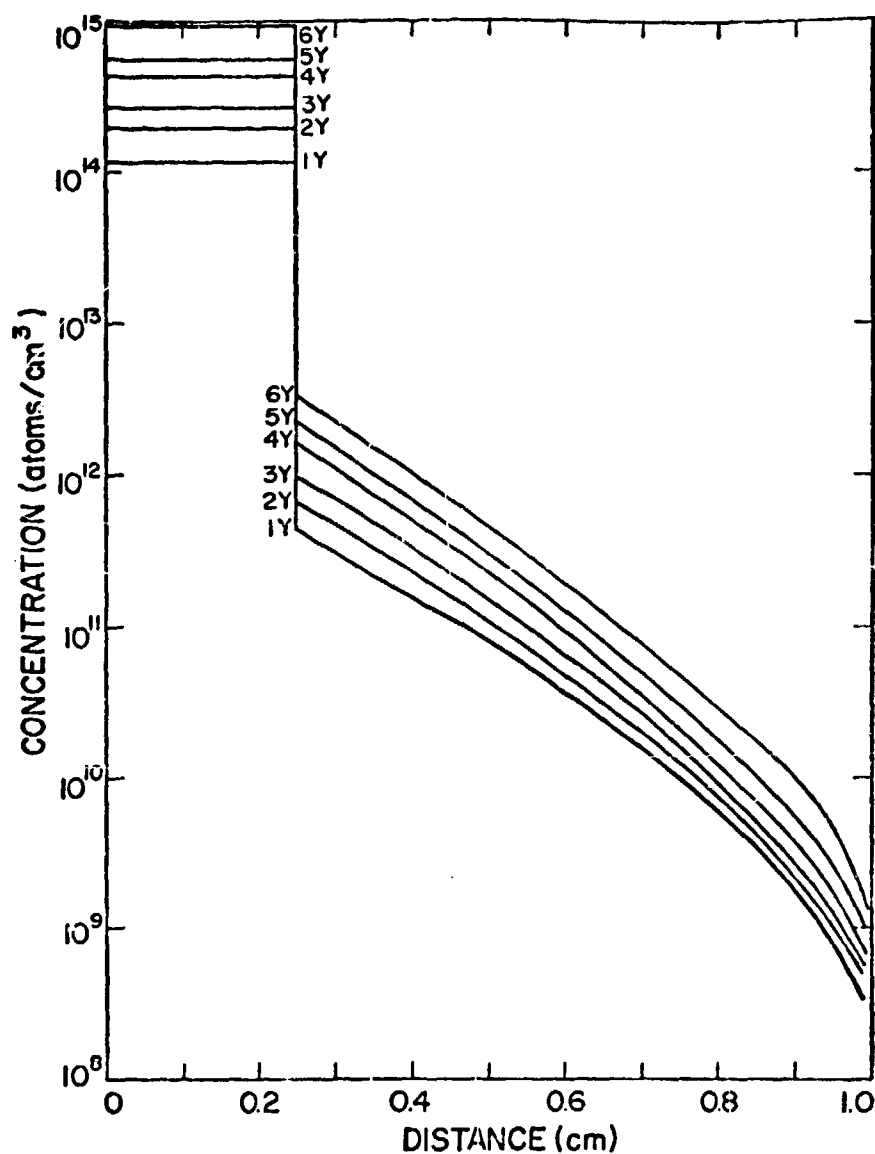


Fig. 3. ^{90}Sr concentration profiles.

amount of ^{90}Sr produced with amount retained in the fuel and structural graphite indicates that about .02% of this species is held up by the presence of the graphite. On the other hand the ^{90}Y does not diffuse significantly but decays into ^{90}Zr .

CONCLUSIONS

Preliminary calculations with the DASH code presented in this paper indicate that DASH is an efficient and accurate code for handling multicomponent diffusion and decay. With diffusion coefficients for the ^{90}Sr species in graphite that are commonly cited, the holdup of the ^{90}Sr in graphite has been shown to be small. The results depend strongly on the diffusion coefficient data, and on the fact that adsorption has been neglected in this analysis.

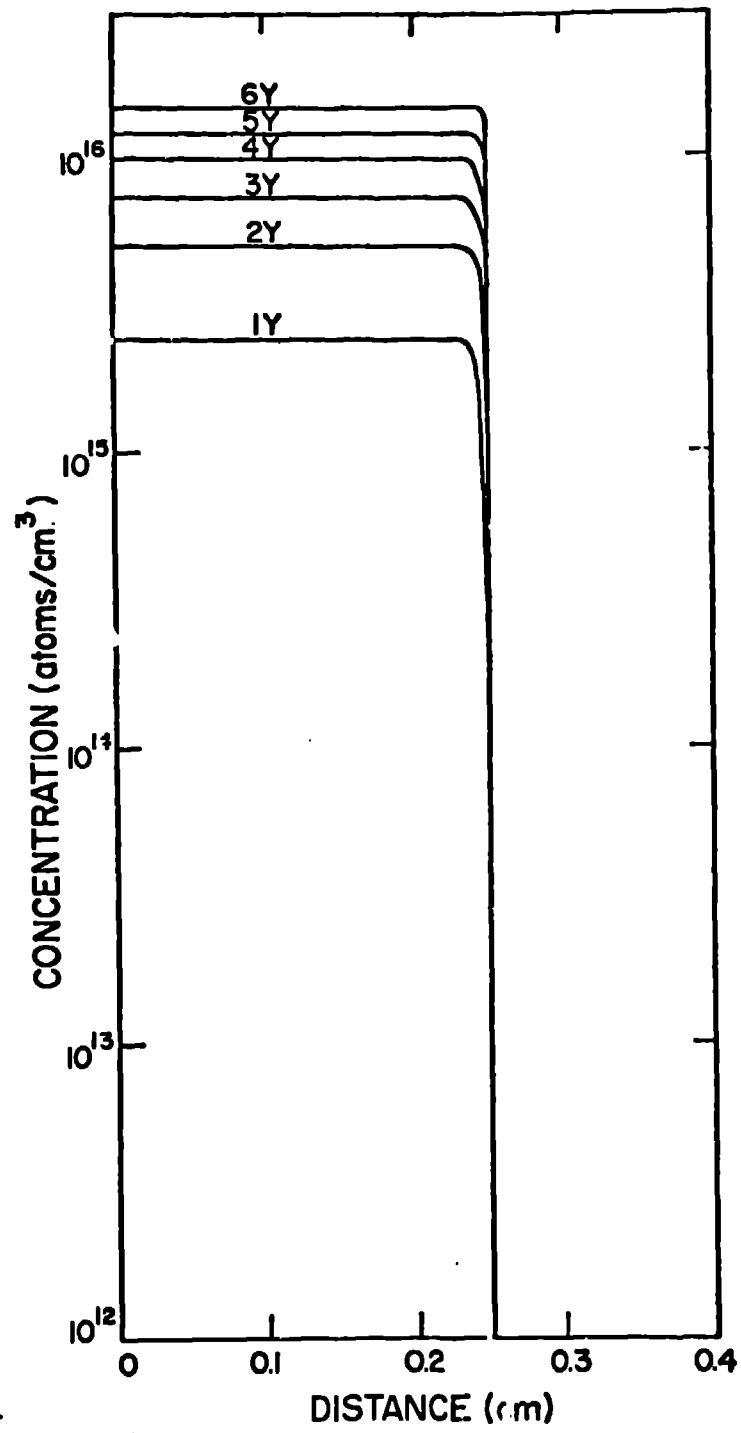


Fig. 4. ^{90}Y concentration profiles.

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